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# **A Versatile Droplet Sizing Spectrometer for Aerosol Hygroscopic Growth Measurements from Research Aircraft**

## **Technical Objectives**

The uptake of water by ambient aerosols at elevated relative humidity (RH) is ubiquitous and often results in large changes in particle size. Because the scattered light intensity from aerosols varies as the diameter taken to the sixth power, optical properties such as the aerosol extinction coefficient, single scatter albedo and optical depth will have strong particle size dependence. There are currently no viable techniques available to perform rapid size-resolved hygroscopic growth measurements over particle size and RH ranges relevant to determining ambient aerosol optical properties at ambient humidity.

The Phase I efforts have focused on establishing the technical feasibility of a new method to measure size-resolved water uptake by aerosols that employs differential mobility analysis (DMA) in-concert with multiple humidification and optical sizing steps. In the technique, monodisperse dry particles selected by a DMA are optically sized by five optical particle counters (OPC), one operating at the same RH as the DMA and the remaining OPCs operating at different controlled RHs so that the particle growth as a function of RH can be observed. The technical objectives of the Phase I included: development of a model of overall instrument response, development of the particle humidification system design, extension of existing thermodynamic models of particle growth, model calculations of the time-dependent growth and evaporation of droplets, implementation and testing of the iterative technique for determining particle dry and wet refractive indices from the simulated OPC response to DMA-selected dry particles, sensitivity studies of the size and refractive index resolution of the OPC, detailed engineering design development of the DMA-OPC systems, and initial bench-top prototyping of the OPC.

In this final report of our Phase I work, we first describe the technical details of the proposed OPC design and then present the major results from our various model simulations of instrument response and the technique for deriving particle index of refraction.

## **Description of Significant Results**

The first major task of the Phase I was to develop a theoretical model of OPC response to be used to optimize the OPC design and to perform sensitivity studies on the anticipated resolution in OPC-measured particle size, refractive index and hygroscopic growth factor. The specifications of the OPC light source, optics, and light gathering systems were required before a detailed model of the OPC response could be constructed. Here we describe the technical details of the OPC, below we present results from the OPC response model calculations. A schematic of the proposed system is shown in Figure 1, the OPCs each operate in parallel and the DMA outlet flow is split between

them. A detailed drawing of the OPC design under consideration is shown in figure 2. For the scattering geometry shown, the elliptical reflector gathers 70% of the full spherical scattering. The transmittance of the optical system, including the heat filter, three lenses and chamber window, is 75%. For a 10 watt white light source,  $9800 \text{ W m}^{-2}$  is delivered to the focal point of the reflector in the 300-650 nm wavelength range, which is the range of sensitivity of the photomultiplier tube (PMT) to be used. Mie calculations of the scattered light energy from 0.1 and 1  $\mu\text{m}$  diameter ammonium sulfate particles indicate that  $5 \times 10^{-9}$  and  $2 \times 10^{-4} \text{ W m}^{-2}$ , respectively, would be scattered and available for collection by the elliptical reflector. We note that the scattered light energy calculated for 0.1  $\mu\text{m}$  particles is larger than but close to the Rayleigh value calculated for scattering by gases. For a reflector area of  $1.1 \times 10^{-3} \text{ m}^2$ , a PMT quantum efficiency of 0.2, and a PMT gain between  $4 \times 10^4$  and  $2 \times 10^3 \text{ Amp/W}$  for 0.1 and 1  $\mu\text{m}$  diameter particles, respectively, an output signal from the PMT between 0.04 and 85  $\mu\text{A}$  is anticipated. A pre-amplification circuit, already available at Brechtel Mfg. Inc. (BMI), will be used to convert the  $\mu\text{A}$  output of the PMT to a 0-10 VDC signal that can be logged by a high-speed digitizer. A similar amplifier circuit will be used with a photo detector to provide real-time monitoring and feedback control of the light source output. The PMT is output current limited to 100  $\mu\text{A}$ , therefore a lower PMT gain may be necessary for larger particles. The PMT dark current (noise) specification is between 0.004 and 0.02  $\mu\text{A}$ ; however, other potential sources of background noise, for example errant light, will need to be stringently controlled to minimize PMT noise. For a particle transit time in the light beam of 1500  $\mu\text{s}$ , between 17 and  $6.7 \times 10^5$  photons/particle will impinge upon the PMT for particle sizes between 0.1 and 1  $\mu\text{m}$ , respectively. The PMT photon dark count over the pulse duration is about 1, however, Rayleigh scattering and other sources of background light will increase the PMT background signal if not properly controlled. Blackened surfaces, proper light trapping, anti-reflection coated optics and other measures will be implemented to reduce background noise. With the aforementioned design features, calculations indicate that the proposed OPC design will be capable of detecting particles in the 0.1 to 1  $\mu\text{m}$  diameter size range.

In addition to the optical sizing consideration described above, we have addressed other OPC design issues, including: sampling time required as a function of particle size and concentration for acceptable Poisson counting error, coincidence of particles in the scattering volume, response to non-spherical particles, and the presence of absorbing material. Table 1 summarizes calculations of the sampling times required for concentration uncertainties less than 20% for 0.3 and 1  $\mu\text{m}$  diameter particles under polluted ( $10,000 \text{ cm}^{-3}$  total number concentration) and clean ( $1,000 \text{ cm}^{-3}$ ) conditions. A Junge power law relation was assumed to model the ambient number size distribution. To estimate sampling times for the listed Poisson concentration uncertainties, charging efficiency and DMA transfer function factors were applied to the assumed ambient concentrations. The dry monodisperse particles were assumed to separate into two major hygroscopic modes each resolved by 10 OPC channels. The noted sampling times are those required to obtain the listed concentration uncertainties in each of the two OPC channels representing the number peaks of the two hygroscopic modes. Approximately 10.3 s are required for particles to flow through the DMA and residence time in the particle humidifiers is about 0.7 s. Therefore, roughly 11 s are required between each

DMA voltage change to allow the newly selected particles to reach each OPC. Based on the above assumptions, the total sampling time to obtain hygroscopic growth factors of 0.3 and 1  $\mu\text{m}$  particles at four different RHs under polluted conditions is approximately 44 s ( $=2 \times 11 + 2 + 20$ , see Table 1). Adding three additional DMA-selected particle sizes smaller than 1 micron would increase the total sampling time, but the total time for all five sizes would still be less than about 80 s, depending on the sizes chosen and the ambient number size distribution. In comparison, currently available techniques would require multiple-hour sampling times to obtain the same data. Due to the much lower concentrations under clean conditions the sampling times are longer to obtain statistically significant samples. Noted DMA sizing uncertainties have been calculated for measured DMA flow rate, voltage, pressure, and temperature uncertainties in airborne DMA systems developed by the PI. OPC sizing uncertainties have been estimated based on response calculations (described below) and are consistent with currently available OPCs. Coincidence of more than one particle within the optical scattering volume at the same time is only a problem for particles smaller than 0.15  $\mu\text{m}$  and total concentrations larger than 10,000  $\text{cm}^{-3}$ . An on-line sample flow dilution system will be used to allow proper operation of the device at total concentrations higher than 10,000  $\text{cm}^{-3}$ . The response to non-spherical particles and the influence of absorbing material on the OPC response are two areas that require additional work. As part of an on-going NSF project led by F. Brechtel to study the optical properties of Asian aerosol, BMI is implementing the Discrete Dipole Scattering code (DDSCAT, *Draine and Flatau* 2000) on a newly purchased Linux workstation dedicated to calculating light extinction by non-spherical particles.

Having evaluated the feasibility of the proposed OPC design with respect to particle sizing and counting, the OPC-response model was extended to examine the OPC sensitivity to refractive index and size changes during simulated particle water uptake measurements. The results from calculations of OPC response for two dry particle sizes and compositions (pure ammonium sulfate and internally mixed ammonium sulfate/soot) are shown in figure 3. Theoretical OPC responses for particles composed of pure ammonium sulfate, soot, and water are shown as the solid, dashed and dotted lines, respectively. Results from the calculations of initially dry particles exposed to increasing RH (85-94%) are shown in each panel of figure 3 as diamonds for 0.3 micron and squares for 1 micron particles. In figure 3a, the response to initially dry 0.3 micron ammonium sulfate particles (diamond) lies on the OPC response curve for pure ammonium sulfate and moves toward the response curve for pure water as it is exposed to increased RH and grows. The results in figure 3 suggest that the OPC will have a monotonic response for particles that grow to sizes smaller than about 1 micron. The ratio of OPC responses to droplets at 88% and 85% is roughly two, with similar ratio values for other RHs and droplets smaller than 1 micron. The large value of the ratio indicates that the OPC will have good hygroscopic growth factor measurement resolution for submicron aerosol. The sizing of larger droplets will be more influenced by the non-monotonic Mie scattering response for larger sizes ( $>0.6 \mu\text{m}$ ). As particles closer to 1 micron are selected, there are ambiguities in OPC sizing, as indicated by the similar OPC response for the 1 micron particles in figure 3a. This feature is typically focused to a small size range, for example, between 1.3-1.6 microns. One of the strengths of the proposed

technique is that OPC sizing ambiguities can be overcome by using the dry index of refraction derived from the dry-DMA/dry-OPC measurement in the solution for the wet droplet sizes that best fit the observed OPC response. This technique is described below. The results in figure 3b provide an example of the sensitivity of OPC response to index of refraction. Significantly lower OPC response is seen for the 0.3 micron internal mixture of ammonium sulfate/soot due to the absorption by soot and the lower growth factors (for the same RHs as in fig. 3a) associated with the lesser soluble material compared to the particles in figure 3a. The monotonic increase in OPC response in figure 3b for the droplets formed on 1 micron particles is much different than the behavior in figure 3a, suggesting that even in the presence of absorbing material there can be good sensitivity of the OPC response to differences in index of refraction similar to those expected in ambient particles.

The second major task of the Phase I work was to develop a design for exposing sampled particles to controlled RH before each OPC. We have developed several humidification systems for ground-based Humidified Tandem Differential Mobility Analyzer (HTDMA) systems; the HTDMA is the traditional technique used to measure particle hygroscopic growth. We will extend our previous designs and use 1/8" diameter nafion tubing reinforced with a stainless steel stiffening mesh to humidify the particle airflow of each OPC. A sheath flow of controlled RH is passed over the outer surface of the nafion membrane and passes into the optical volume of each OPC to maintain a constant RH over the droplets through the entire optical sizing process. The purpose of the stiffening mesh is to provide structural support to the nafion and prevent its collapse during flight operations. The controlled-RH sheath flow is produced by mixing dry and saturated air. The desired RH is maintained by controlling the ratio of dry-to-wet flow rates with solenoid valves and a feedback loop with a RH sensor.

In order to simulate the instrument response to aerosol water uptake in the sample humidifiers, we must be able to model the hygroscopic growth of laboratory and ambient particles of various compositions and sizes. The third major task of the Phase I effort was to adapt existing thermodynamic models of particle water uptake (*Brechtl and Kreidenweis* 2000a; 2000b) to compositions more representative of ambient aerosols. The new version of the thermodynamic model includes major organic acids (malonic, succinic, glutaric, propanoic, oxalic) found in the ambient aerosol in addition to the major inorganic salts ( $(\text{NH}_4)_2\text{SO}_4$ , NaCl,  $\text{NH}_4\text{NO}_3$ ,  $\text{NH}_4\text{HSO}_4$ ) from the original model. In addition to treating particles composed of single chemical species, the model is capable of calculating the droplet size at a given RH for internal mixtures of the above compounds. Results from the growth model for an internally mixed ammonium sulfate-glutaric acid particle are shown in figure 4. Growth factors for pure ammonium sulfate are shown in the figure for comparison purposes and the 'x' symbols summarize observed growth factors from numerous HTDMA field studies on ambient aerosols. The modeled growth results for the internally mixed composition better simulates observed growth of ambient particles than the results for pure ammonium sulfate. The model results agree well with other reported growth results for organic acids and reproduce the observed growth of internally mixed ammonium sulfate-secondary organic aerosol formed during smog chamber experiments. The growth model developed during Phase I

activities will play an important role in validating OPC response during future laboratory calibrations by providing calculated droplet sizes against which measured OPC and HTDMA droplet distributions may be checked. The growth model will also help establish connections between observed growth and chemical composition for ambient measurements where particle composition may or may not be known.

A second component of the Phase I hygroscopic growth modeling effort and the fourth major task of the Phase I work was the development of a new time-dependent model of droplet diffusion growth and evaporation to investigate the sensitivity of droplet size to variations in RH within the instrument. Changes in droplet size during the measurement can occur due to RH and temperature gradients in the instrument and are especially a concern when droplets exit a humidifier and are transported through the OPC. To investigate the sensitivity of droplet size to variations of RH within the instrument, the droplet diameter adjustment times have been calculated as a function of droplet size and changes in the RH of the air surrounding the droplet. Chemical composition may also play an important role in determining adjustment times as some organic compounds may require a relatively long time to reach their equilibrium size compared to inorganic salts. Chemical composition effects require further study. For an assumed  $\pm 2\%$  RH variation within the instrument, calculations indicate that changes in droplet diameter of between  $\pm 1$  and 5% can occur for 0.1-1 micron droplets over the timescales of transport through the OPC ( $\sim 1$ -3 ms). To maintain changes in droplet diameter of less than 1-2% the RH inside the OPC must be kept within 1% of the humidification system set point. This stringent design constraint will be addressed through precise temperature control of each humidifier/OPC pair and by surrounding the aerosol sample stream entering each OPC with a humidified sheath flow.

Another major task of the Phase I effort was the development of an overall model to simulate the instrument response to ambient aerosols of various sizes and compositions that included DMA size-selection, particle hygroscopic growth in the humidifiers, and optical sizing and counting. The models of OPC response and particle hygroscopic growth described above were integrated with a model of DMA size-selection so that the entire measurement sequence could be simulated. The presence of larger, multiply charged particles (up to three charges) in the DMA monodisperse sample flow was included in the DMA size-selection model and the OPC response to these particles examined. The integrated model of instrument response was used to test design concepts and investigate measurement sensitivities. Figure 5 shows an example of model-simulated selection of dry monodisperse 313 nm diameter singly-charged and larger diameter multiply-charged ammonium sulfate particles with the DMA, the response of the OPC to the dry particles (dashed line), and the response of the OPC to the hydrated aerosol at four RHs between 84% and 96% (gradually increasing line thickness). The results shown were obtained using Mie theory to determine the scattered light energy perpendicular and parallel to the scattered plane as a function of particle size for a single particle. The modeled response was then scaled according to the particle count in the given size bin and normalized to the maximum DMA-selected singly charged particle count. It is assumed in the current model that particle composition does not vary with particle size, which is not the case for ambient aerosol. Under the assumption of size-

independent composition, all particles have the same refractive index and grow equally when exposed to a given RH. Results from other simulations that more realistically simulated the hygroscopic growth of ambient aerosol were qualitatively similar to those shown in figure 5. The simulations show that as the droplets grow at higher RH, the increase in droplet diameter increases the modeled scattering more than the reduction in droplet refractive index decreases the scattering, consistent with the results in figure 3. As water is added to the droplets, the index of refraction approaches that of water (1.3), which is smaller than that for ammonium sulfate (1.5). The model of overall instrument response provided a framework for simulating the entire measurement process and allowed us to test the iterative procedure (described below) for determining particle refractive index from the OPC measurements.

A major benefit of the proposed DMA-OPC measurement system is that refractive indices can be derived for sampled dry and wet particles. The fifth major task completed during the Phase I was the development of the iterative technique using simulated OPC calibration equations as a function of refractive index to derive a value for the dry particle and humidified droplet refractive indices consistent with the observed dry mobility and dry or wet optical volume and/or number size distributions. Each OPC calibration equation describes the OPC response as a function of particle size for a constant refractive index. A set of OPC calibration equations is required as input to the iterative procedure and describes the OPC response as a function of size for a range of refractive indices representative of the ambient aerosol (e.g. 1.3-1.9). The procedure employed is similar to that used by *Hand and Kreidenweis* (2002). The simulated OPC calibrations used currently will be replaced with actual calibration equations when laboratory characterization studies of the present OPC design have been completed. If simultaneous size-resolved composition information is available, that information will also be used, but is not necessary, as a constraint in determining the particle index of refraction. In the iterative approach the ‘dry’ refractive index is determined that minimizes the difference between the observed DMA and OPC dry volume or number size distributions. A range of dry particle refractive index values could be derived depending on the chemical heterogeneity of the monodisperse particles selected by the DMA and the sensitivity of the OPC sizing to the differences in composition. An example of the application of the iterative procedure to model-simulated volume distributions of dry 146 nm DMA-selected ammonium sulfate particles is shown in figure 6. The DMA volume distribution is shown as the solid line while the ‘as-observed’ unadjusted OPC volume is shown dotted. Using the calibration equations of *Hand and Kreidenweis* (2002), the iterative procedure was applied to determine a dry refractive index and associated adjusted OPC volume distribution (dashed line in figure 6) that agreed best with the DMA volume distribution. The variation of the Chi-squared difference between the DMA and OPC volume distributions as a function of refractive index is shown in figure 7. As shown in figure 7, for this example calculation the smallest Chi-squared value was found for a refractive index of 1.54, which compares favorably to the value of 1.53 for ammonium sulfate. In principle, the iterative procedure could be performed separately on the multiply charged particles, shown in figure 6 by the increasing concentrations at larger sizes, allowing refractive indices to be derived for larger diameter particles. Future versions of the iterative procedure will include this



capability. The results of our tests of the iterative procedure to determine dry particle refractive index indicate that derived values agree within 2-3% of ‘true’ values.

Once the dry state is determined, the same iterative procedure described above is applied to the wet OPC measurements at a given RH using the dry particle size and derived dry indices of refraction as inputs to describe the dry particle state. During OPC calibration studies on droplets, the iterative procedure is used to determine the refractive index corresponding to the best agreement between calculated and measured OPC droplet distributions. For droplets of known composition, the adjusted OPC distribution is also compared to the calculated droplet distribution at the given RH for the dry particle size and composition selected by the DMA. For ambient OPC hygroscopic growth measurements, the iterative procedure assumes different hygroscopic growth factor distributions and calculates corresponding values of droplet refractive indices using volume-mixing rules and the derived value of the dry index of refraction. The OPC calibration equations are then used as above to calculate the OPC droplet number or volume distribution to be compared with the measured OPC droplet size distribution. The growth factor distribution that minimizes the difference between calculated and measured OPC distributions is chosen as the solution. The derived distributions of growth factors from OPCs operating at lower RHs will serve as additional constraints on growth factor distributions for OPCs operating at higher RH. Figure 8 shows results from the application of the iterative technique to the above example of DMA-selected dry 146 nm ammonium sulfate particles exposed to 84% RH. The format of the figure is similar to figure 6 above, the solid line designates the ‘true’ growth determined by thermodynamic calculations, the dashed line designates the retrieved OPC wet distribution using the iterative procedure, and the dotted line designates the unadjusted OPC wet distribution. The solid line in figure 8 would be the growth measured by a second DMA operated at 84% RH (e.g. in an HTDMA system) and sampling the dry 146 nm particles. The Chi-squared values returned by the iterative procedure for different values of refractive index are shown in figure 9. The theoretical value of the droplet refractive index for this case is 1.384 and a value of 1.37 was retrieved by the iterative procedure, indicating an error of roughly 2%. The theoretical droplet growth factor for ammonium sulfate particles at 84% RH is 1.56 (droplet diameter of 226 nm), the retrieved OPC wet distribution is peaked at slightly smaller sizes (220 nm) compared to the ‘true’ droplet distribution but the agreement is still within 3%. Results from tests of the iterative procedure to determine droplet distributions in the OPC indicate that the proposed technique is capable of retrieving droplet refractive indices and hygroscopic growth factors within 3-7% of their ‘true’ values.

The final major task of the Phase I effort was to use the results from the above modeling and sensitivity studies to develop the instrument prototype schematics and specifications. Engineering assembly drawings of most of the major instrument systems are shown in figures 2 and 10. A summary is provided in Table 2 of the major components with estimated physical dimensions, weight, and power specifications. The instrument design has been optimized for deployment on the CIRPAS Twin Otter aircraft; therefore special consideration has been given to minimize the overall size, weight and power requirements of the system. Relative humidity, temperature, and

pressure must be monitored throughout the system to ensure stable operation, especially with regard to the strong sensitivity of RH to variations in instrument temperature. We have operated systems capable of controlling flow RHs to within  $\pm 0.9\%$  RH of set point values with a nominal sensor precision of  $\pm 0.8\%$  (Brechtel, 1998). An on-line calibration system employing an aerosol atomizer with a salt solution containing polystyrene latex spheres will be integrated into the hygroscopic growth system and used to challenge the instrument with particles of known size and composition during research flights. In order to validate and improve the OPC response model, BMI has initiated construction of a prototype OPC. The optical system and OPC chamber have been completed and the light-pulse detection electronics are near completion. Once assembled, the OPC will be challenged with DMA-selected laboratory aerosol to examine its response characteristics.

In summary, we have developed a detailed design for an OPC and used the optical geometry and other specifications in a model of OPC response to show that particles in the 0.1-1  $\mu\text{m}$  size range should be successfully detected. New thermodynamic and kinetic models of particle hygroscopic growth were constructed and used as part of a model of overall instrument response to simulate the growth of particles in the humidification system and to examine the potential for transient droplet diameter changes due to small changes in instrument RH. The proposed iterative procedure to determine hygroscopic growth factors and refractive indices was tested and the agreement between calculated and 'true' values was between 3 and 7%. Overall, the modeling studies indicate that the proposed new instrument should be capable of observing hygroscopic growth factors at multiple sizes and RHs with a time resolution two orders of magnitude better than existing techniques, making it appropriate for aircraft operations.

**Prepared by:**

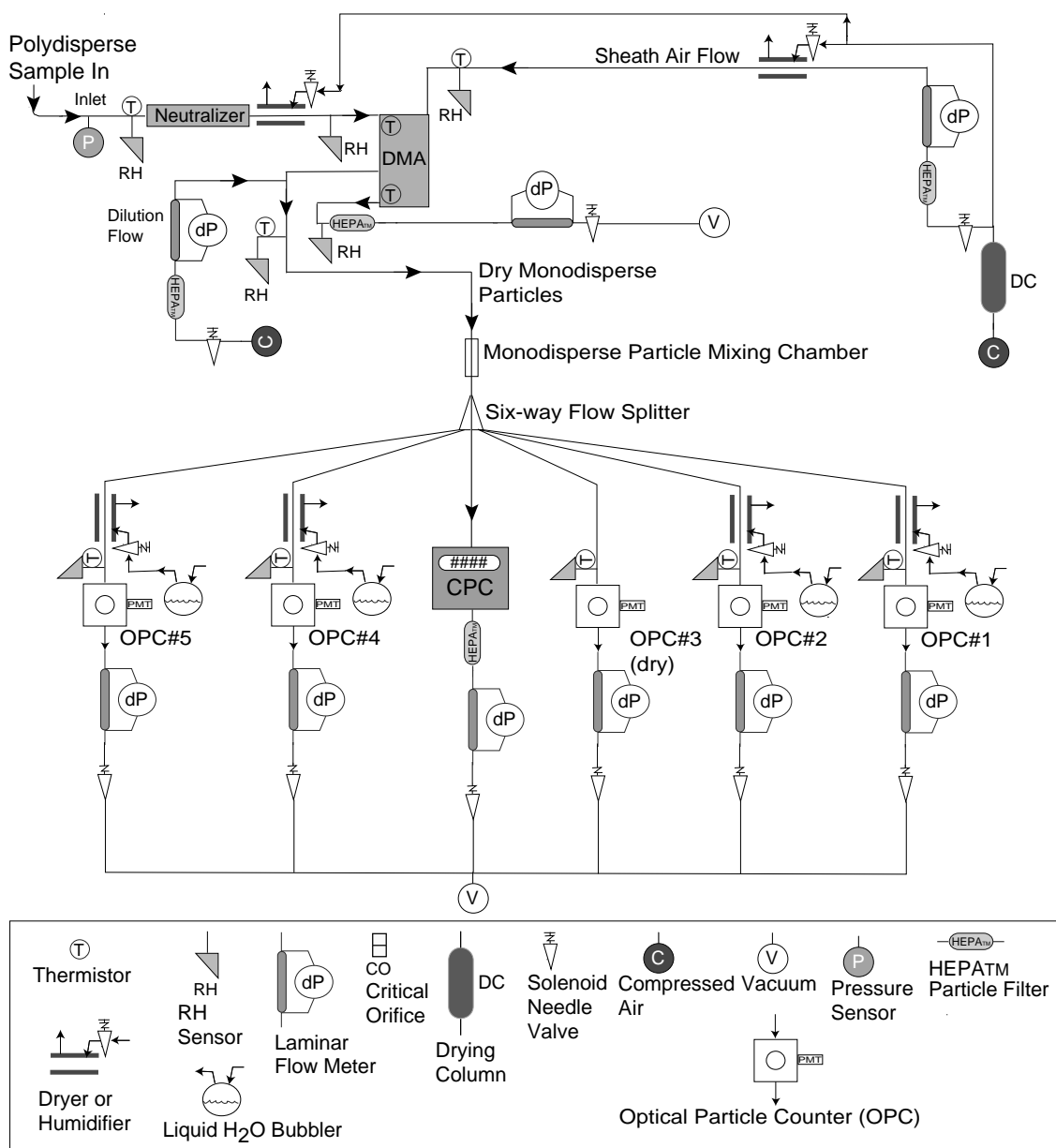
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**Table 1 Summary of estimated sampling times, and concentration, sizing and hygroscopic growth factor uncertainties for clean and polluted sampling conditions.  $N_{\text{ambient}}$  denotes the peak monodisperse number concentration in each OPC hygroscopic mode. Sampling time does not include 11 s transit time of particle through DMA and humidifier after a voltage change in the DMA.**

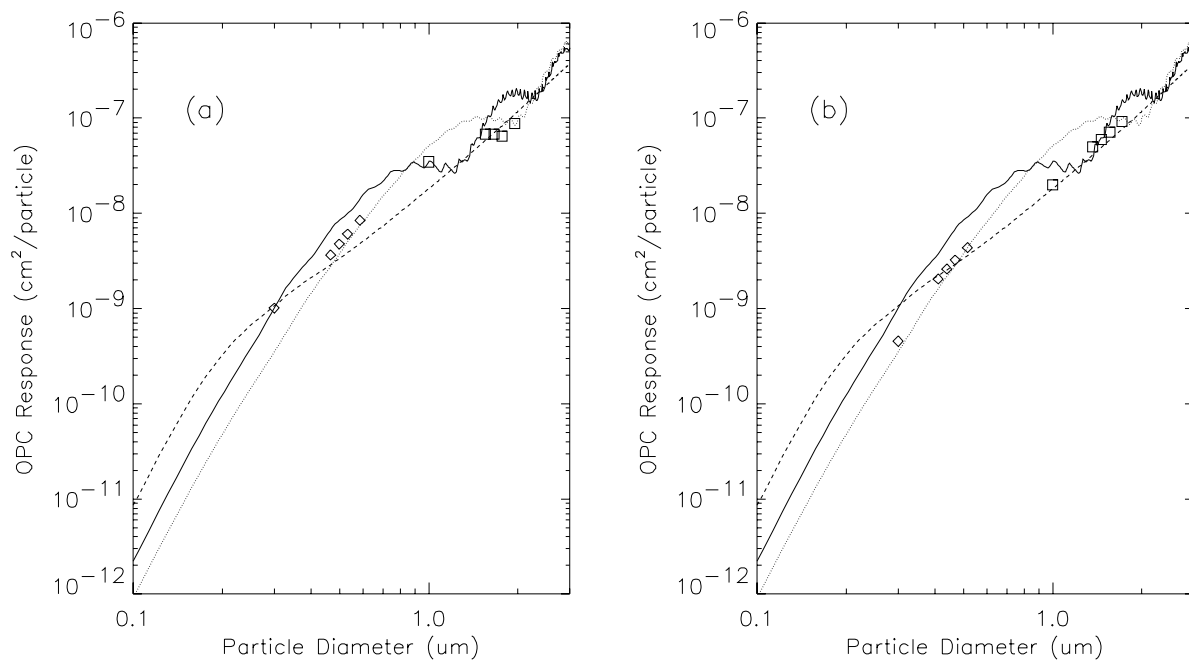
Case	$N_{\text{ambient}}$ ( $\text{cm}^{-3}$ )	Sampling Time (sec)	Concentration Uncertainty (+/-%)	DMA Size Uncertainty (+/-%)	OPC Size Uncertainty (+/-%)	Growth Factor Uncertainty (+/-%)
0.3 $\mu\text{m}$ Polluted	13.6	2	15	3	5	6
1 $\mu\text{m}$ Polluted	0.5	20	20	3	7.5	8
0.3 $\mu\text{m}$ Clean Marine	1.4	18	15	3	5	6
1 $\mu\text{m}$ Clean Marine	0.05	200	20	3	7.5	8

**Table 2 Summary of preliminary physical dimensions, weight and power specifications of the proposed instrumentation. ‘BMI’ designates components built by Brechtel Mfg. Inc.**

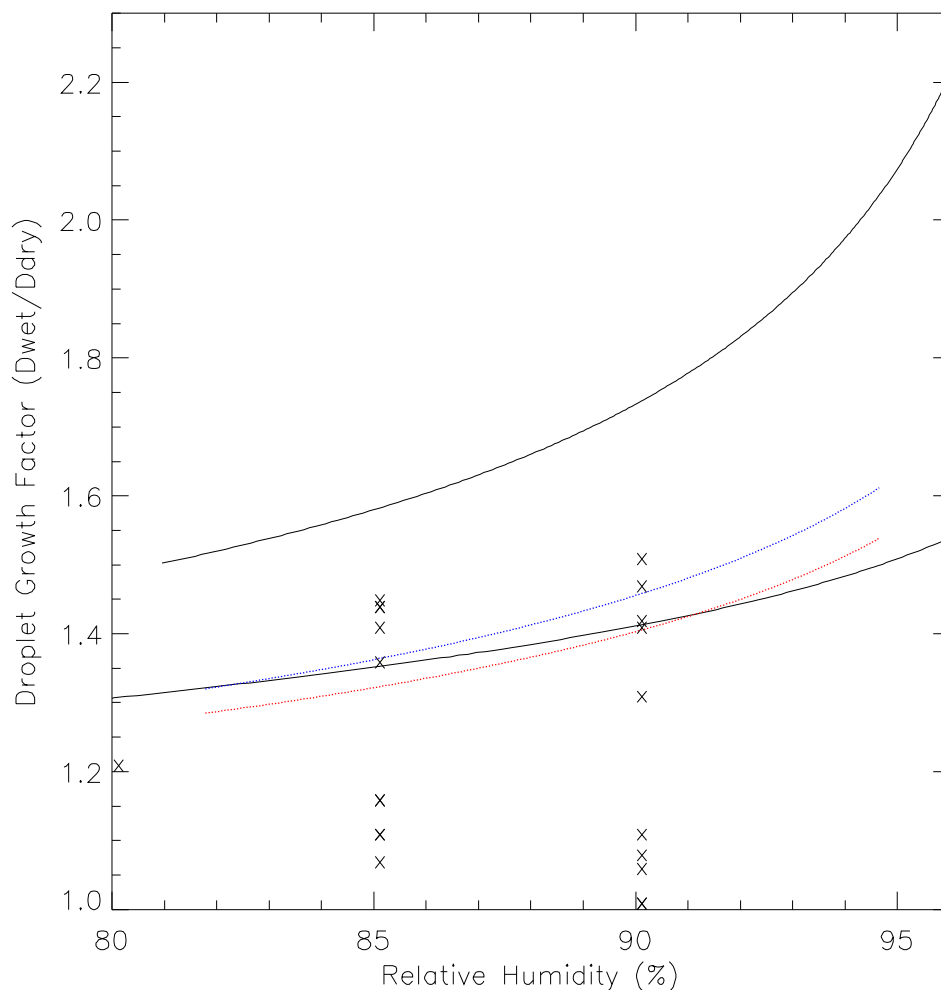
Component	Description or Model	Weight (lb)	Dimensions (LxWxH) (in)	Power (Watts)
Sample/DMA Sheath Air Humidifier	Permapure (PD-50T-SS); BMI	3	16x1.25x1.25	20
Humidification Control	BMI	10	6x6x4	40
Charge Neutralizer	BMI	6	8x2x2	0
Sample/DMA Air Dryer	BMI	11	12x6x3	25
Hi-Flow DMA + Cal. System	BMI	15	24x5x5	0
DMA High Voltage Supply	Bertan	1	5x2x1	4
Optical Particle Counters (x5)	BMI	3 each	2x2x4	62
Condensation Particle Counter	TSI Model 3010	12	8x8x8	25
DMA Flow Control System	BMI	8	8x6x3	30
P, T, RH sensors	Various	5	8x8x2	6
Data Acquisition System	PC; Display; A/D	21	10x8x6	105
Misc. Mechanical	Fittings/Tubing	15	6x6x6	0
Misc. Electronics	DC Power Supplies	10	8x4x6	25
DC Vacuum Pump	GAST 2032	11	6x5x12	90
Instrument Mechanical Structure	Techmar 19" wide; 24" dp; 36" tall single-wide rack mount chassis	20	24x23x36	0
<b>TOTAL</b>		<b>163</b>	<b>24x23x36</b>	<b>432</b>



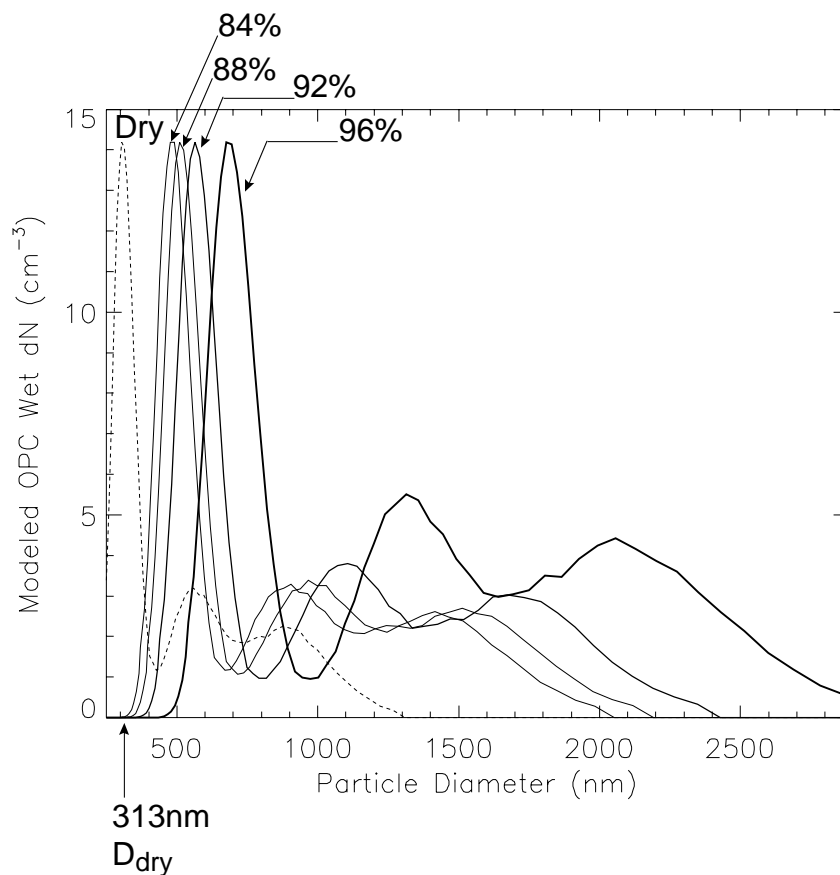




**Figure 3.** Results of Mie calculations for initially dry 0.3  $\mu\text{m}$  (diamonds) and 1  $\mu\text{m}$  (squares) particles composed of (a) pure ammonium sulfate, and (b) internally mixed ammonium sulfate and soot. Solid lines designate calculated OPC response to pure ammonium sulfate particles; dashed lines are OPC response to pure soot particles, and dotted lines represent OPC response to pure water droplets.

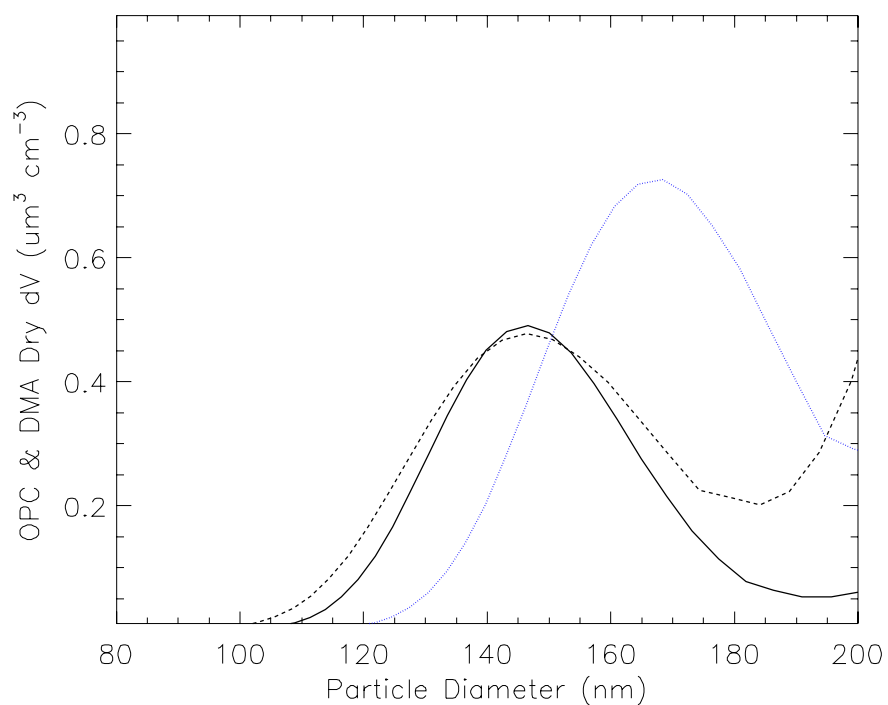


**Figure 4.** Model calculations (this work) of the equilibrium growth of an initially dry  $0.18\ \mu\text{m}$  diameter pure ammonium sulfate particle (upper solid line) and an internally-mixed particle of the same diameter composed of 21% by mass ammonium sulfate and 79% by mass glutaric acid (lower solid line). Upper dashed line denotes *Peng et al.* (2001) parameterization of the growth of pure glutaric acid containing particles. Bottom dashed line denotes unpublished HTDMA growth measurements on secondary organic aerosol formed on ammonium sulfate seed particles during oxidation of cyclohexane by ozone in the Caltech smog chamber. 'X's denote ambient HTDMA results from numerous field studies described in Table 1 of *Brechtel and Kreidenweis* (2000a).

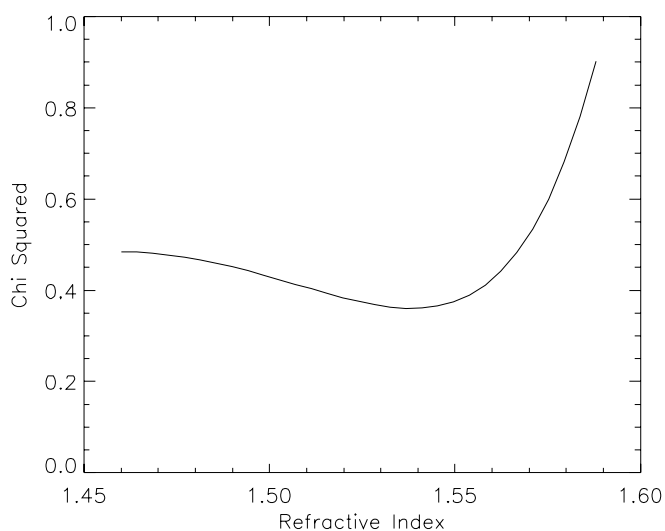


**Figure 5.** Model-simulated singly, doubly and triply-charged DMA particle selection and OPC response to dry (dashed line) and hydrated aerosol (gradually increasing line thickness) as a function of particle size. Initially dry 313 nm diameter singly-charged particles were selected by the DMA.

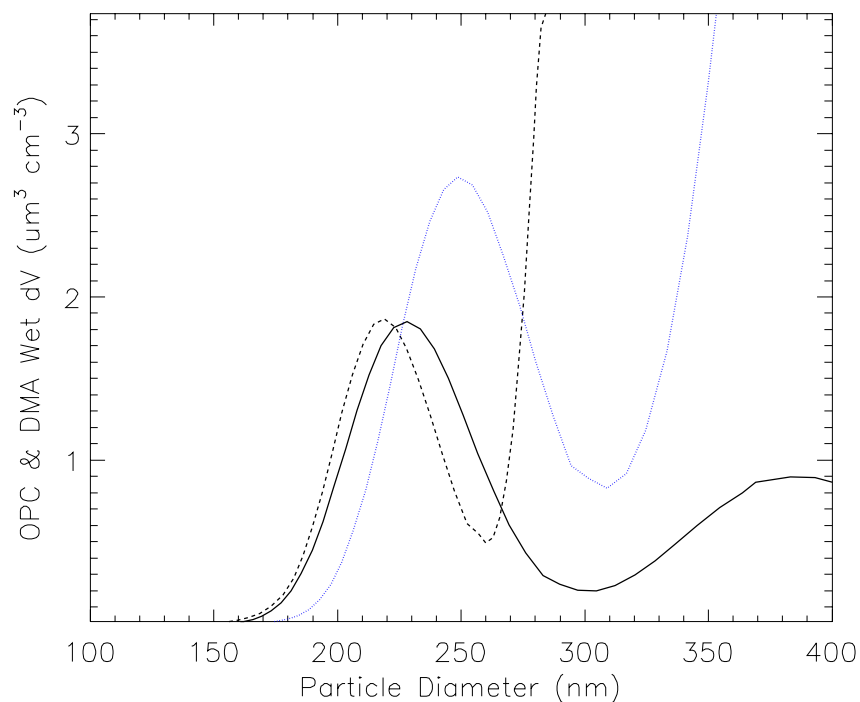




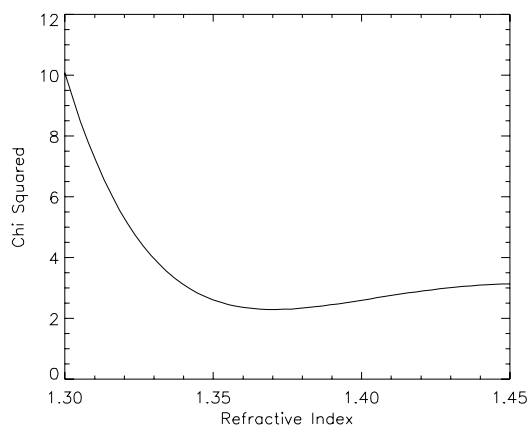
**Figure 6.** Model-simulated volume distributions of 146 nm mobility diameter singly charged DMA-selected particles (solid), unadjusted OPC observed volume (dotted), and adjusted OPC volume (dashed). The iterative procedure to determine the best-fit OPC volume distribution returned a derived refractive index of 1.54.



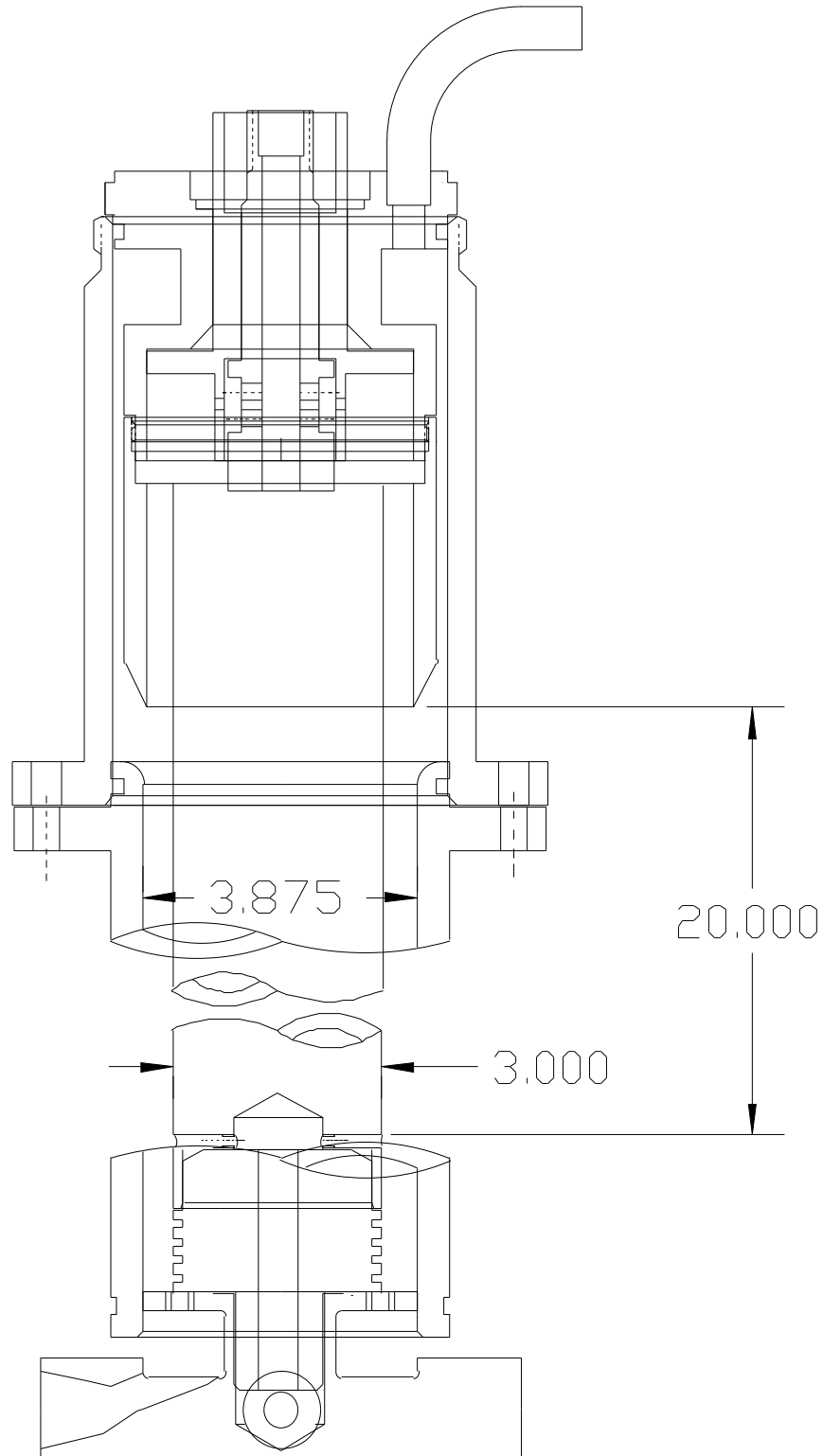
**Figure 7.** Chi-squared difference between DMA and OPC volume distributions as a function of refractive index for the selection of 146 nm diameter particles in Fig. 6. The minimum Chi-squared value was found for a refractive index of 1.54.



**Figure 8.** Model-simulated wet volume distributions of deliquesced 146 nm mobility diameter singly charged DMA-selected particles (solid), unadjusted OPC observed wet volume (dotted), and adjusted OPC volume (dashed). The iterative procedure to determine the best-fit OPC volume distribution returned a derived refractive index of 1.37, within 1.7% of the theoretical value of 1.384. The hygroscopic growth factor determined from the adjusted OPC distribution agrees with the theoretical value within 3%.



**Figure 9.** Chi-squared difference between DMA and OPC wet volume distributions in preceding figure as a function of refractive index for the results in Fig. 8. The minimum Chi-squared value was found for a refractive index of 1.37.



**Figure 10.** Assembly drawing of BMI High-Flow Differential Mobility analyzer showing 3" OD high voltage center rod, 3.875" ID outer cylinder, and 20" effective DMA column length. The design allows selection of 1 micron diameter particles with an applied voltage of 10 kV and sheath air and aerosol flow rates of 8 and 1 lpm, respectively.

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